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Instrumented indentation at elevated temperatures for determination of material properties of fusion relevant materials

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ABSTRACT

The testing of small sized samples is an important advantage of the instrumented indentation with respect to the investigation of materials for fusion application. A continuous recording of the indentation depth and force enables a determination of mechanical properties of the tested material.

In this study, the results of the high temperature experiments with a custom made indentation device are presented. The reduced activation ferritic martensitic steel EUROFER is investigated in an unirradiated state with spherical tips and for the first time Vickers tips at increasing temperatures up to 500 °C.

The indentation procedure is numerically simulated at different temperatures and the corresponding load-displacement-data are compared with the experimental results. A quantification of the influence of variations of the indentation tip radius is presented as well.

Finally, the operation of the indentation device with respect to the restrictions of the Hot Cell environment is discussed.

1. Introduction

The development and qualification of new compositions of structural materials for the application in future fusion power plants is an important field of current fusion research. Hence, the comprehensive characterization of such materials at their operation conditions is an indispensable part of the work of fusion material scientists all over the world. For the investigation of neutron-irradiated materials, the instrumented indentation is an attractive and promising method. The use of small sized indentation samples has a positive effect on the costs of irradiation programs and the dose rate of the single specimen and is indispensable for a future neutron source, because of its small irradiation chamber. With respect to the Hot Cell environment, the relatively simple preparation of the samples simplifies a multiple testing. Therefore, a maximum of information can be obtained from the investigated material.

Instrumented Indentation at elevated temperatures is an important research field with remarkable progresses in the recent years. For example the evaluation of welded materials, nanoindentation at high temperatures, and the investigation of different structures and materials are part of current research, e.g. [1–4]. Especially with respect to investigations related to nuclear fusion, successful work has been done with high temperature indentation experiments [5] and investigations of ion implanted thin layers using nanoindentation [6, 7]. A further possible application could be the investigation of materials with respect to the ductile-to-brittle transition behavior [8–10].

In a fusion reactor, the structural materials have to withstand extreme loads due to the nuclear fusion process, like high neutron radiation and high temperature conditions. A candidate material like EUROFER, a low activation steel, needs to be investigated in irradiated state at the operation conditions of nuclear fusion, to obtain a complete understanding of the material behavior [11].

By using a commercial indentation system at the Fusion Material Laboratory (FML), promising results already were obtained by instrumented indentation experiments at room temperature on irradiated specimens [12].

For investigations at elevated temperatures, a new indentation device was developed at the Karlsruhe Institute of Technology (KIT). In contrast to commercial systems, the device is designed for future remote-handled investigations of neutron irradiated materials in a Hot Cell of the Fusion Materials Laboratory at KIT. The machine enables indentation experiments at temperatures up to 650 °C with a maximal testing force of 200 N [13, 14]. First results of high temperature indentation tests are shown in [15].

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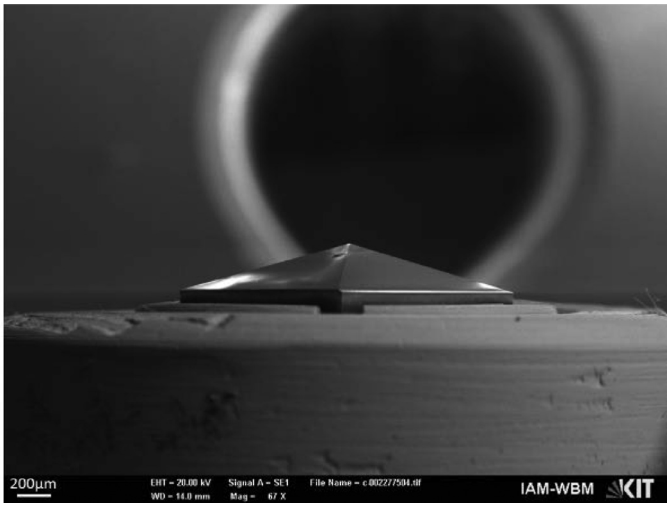

Fig. 1. SEM micrograph of an indenter consisting of a TZM holder and a Vickers diamond tip.

2. Experimental

2.1. Indentation device

The indentation column with the integrated indenter, the sample stage and the heating system are installed inside a vacuum chamber to prevent an oxidation of indenter and sample. The indentation depth measurement system is located outside of the vacuum chamber. The indentation experiments are performed in force control.

The sample and the indenter tip are heated up separately via independent heating cartridges, which are located inside the indentation column and the sample stage. The active temperature regulation uses the signals of a pair of thermocouples. The high temperatures up to 650 °C necessitate a water cooling system. Hence, stable thermal conditions are achieved for the sample and the indenter tip.

For the measurement of the indentation depth, an optical system is used, because of its insensibility to the temperature conditions inside the vacuum chamber. Using image capturing, via a camera in combination with a long distance microscope, and image correlation in post-process, it is possible to determine the indentation depth from the relative movement of the sample and the indenter. The two measurement points of the optical system are located very close to the contact point of tip and sample. Hence, the influence of thermal expansion on the measurement is minimized.

In combination with the force measurement, the load-displacement-curves could be determined for every indentation experiment by using a custom made software with a resolution of 0.1 µm and 0.05 N for the displacement and the force, respectively.

For the use of the high temperature device, custom made indenters are necessary. These indenters consist of a titan zirconium molybdenum (TZM) holder and the tip. In the present study, indenter with rounded Rockwell cones with a radius of 200 µm and pyramidal Vickers tips are used, see Fig. 1. Diamond and sapphire are used as tip materials. A mechanical clamping system fixes the diamond tip in the holder. The pressure sensitivity of sapphire does not allow the use of this system for the sapphire tips. Therefore, an aluminum nitride ceramic adhesive with an application temperature of 3000 °C is used for the fixing of the tip.

All indenters are designed in a way that they can be handled with manipulators in a Hot Cell to enable the use of different tip geometries and the exchange of broken indenters.

More detailed insight in the indentation device and the different setups are given in [14] and [15].

2.2. Material and samples

The tested material EUROFER (9CrWVTa), a customized ferritic martensitic steel, is a specially developed alloy for fusion application. The reduced activation of the material is achieved by substitution of high activation alloy components, e.g. Mo, Nb and Ni, by elements with lower activation, e.g. W, V and Ta, [16]. The heat treatment of the material was 1040 °C for 0.5 h and 760 °C for 1.5 h.

As specimens, broken halves of Charpy impact or mini fracture mechanics tests with a size up to $15 \times 6 \times 3$ mm³ have been used for the indentation experiments. As preparation, the samples were multi-level grinded and subsequently polished with a finish of 3 µm diamond paste.

The indentation experiments are evaluated according to Brinell and Vickers on basis of an optical measurement of the diameter of the indents via an optical microscope according to Brinell and Vickers, DIN EN ISO 6507 and DIN EN ISO 6507.

Additionally, the load-displacement-curves of the individual experiments are determined and evaluated, according to the standard DIN EN ISO 14577. Mono cyclic experiments with a maximal load of 40 N, a loading rate of 1 N/s and a holding time of 15 s were carried out.

All the experiments were carried out between room temperature and 500 °C with spherical and Vickers indenters with diamond and sapphire as tip materials. The indentation depth of all presented experiments exceed the threshold value of 6 µm stated in DIN EN ISO 1477.

The upper temperature threshold for diamond is 400 °C, because of its increasing chemical instability in contact with steel at elevated temperatures. Further information of indenter tip material behavior at elevated temperatures is given in [17]. The vacuum pressure during the experiments was between 8×10^{-6} mbar to 1×10^{-5} mbar.

Due to the limited sample size and the wide range of test temperatures, a multiple indentation at every test temperature was not possible.

3. Results and discussion

3.1. Brinell and Vickers hardness

In this section, the conventional hardness according to Vickers and Brinell are evaluated. In Fig 2, the Brinell hardness (HB), the Vickers hardness (HV) and the ultimate tensile strength (UTS) [11] are plotted vs. the temperature. The results for both sapphire tip geometries exhibit the same temperature dependency of the material hardness of EUROFER and can be verified with the tensile test results (UTS). The hardness decreases continuously with an increasing test temperature until 400 °C. Between 400 °C and 500 °C, the drop of the hardness is stronger.

A comparison with the results of the investigations of MANET II in [15], which were carried out with a diamond tip, shows the practicability, on the one hand of sapphire as tip material and on the other hand of the whole design of the custom made indenters for different tip geometries. Indenter with cube corner tips are already produced and corresponding investigations are planned.

In Fig. 3, the HV of EUROFER is plotted for the diamond and the sapphire tip and a clear variation of the hardness values is visible over the whole testing temperature range. The results determined with the sapphire tip are about 1.5% higher than those obtained with the diamond tip. A similar phenomenon was discovered in a measurement campaign on MANET II with spherical indenter tips, see [15].

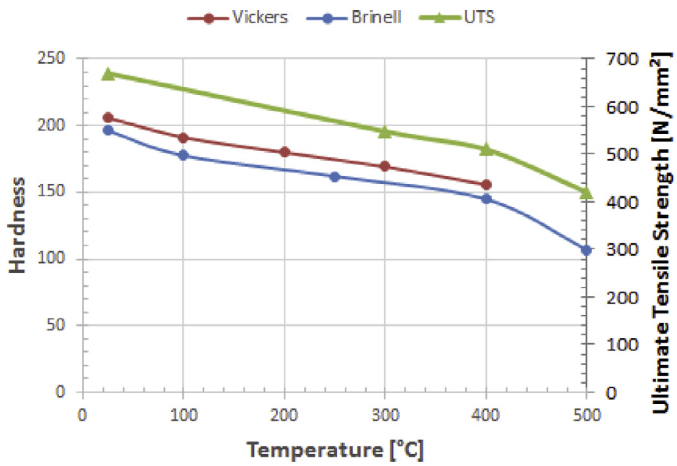

Fig. 2. Brinell and Vickers hardness HB and HV, respectively, of EUROFER, as determined using the sapphire tips compared to tensile testing data from [11].

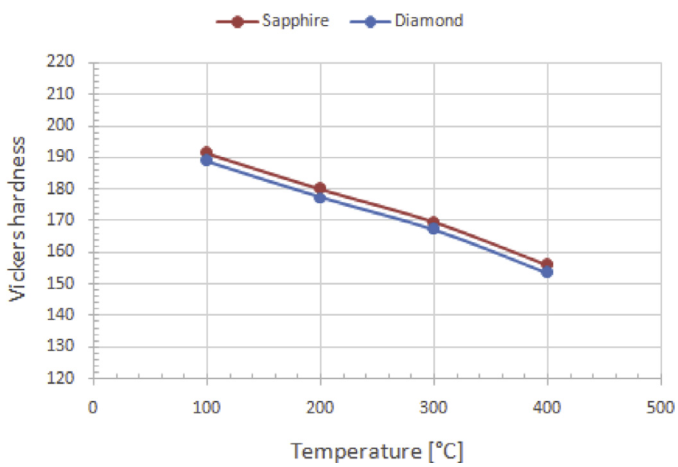

Fig. 3. Comparison of the Vickers hardness determined with the sapphire and the diamond tip for EUROFER.

The fact, that this phenomenon is visible for different indenter tip geometries, confirms the argument of [15], that the elastic modulus of the indenter tip material has a not negligible influence on the evaluated hardness values HB and HV. This fact is investigated via numerical simulation in chapter 4.

3.2. Load-displacement-curves

In Fig. 4, the load-displacement-curves for EUROFER at different testing temperatures are shown for a spherical diamond tip. It is obvious, that the material behavior is strongly depending on the test temperature. The indentation depth increases with an increasing temperature, caused by lower deformation resistance at high temperature. In contrast, the unloading part of the curves seems almost identical for all temperatures. However, a small decrease in the slope of the unloading was observed, which implies a decrease of the Youngś modulus with increasing temperature.

In Fig. 5, the load-displacement-curves for the spherical sapphire tip are shown. A similar material behavior compared to Fig. 4 is observed; an increasing test temperature leads to an increasing indentation depth. In general, the indentation depths with the sapphire tips are lower than for the diamond tip. This is mostly due to a geometrical deviation of the sapphire tip compared to the diamond tip caused by the limited tolerances of the tip fabrication. Therefore, a verification of geometrical deviations via SEM measurements and tests on reference materials are necessary. The elastic behavior of the tip material has an influence on the measured indentation depth as well. This issue is investigated via numerical simulation, compare chapter 4.

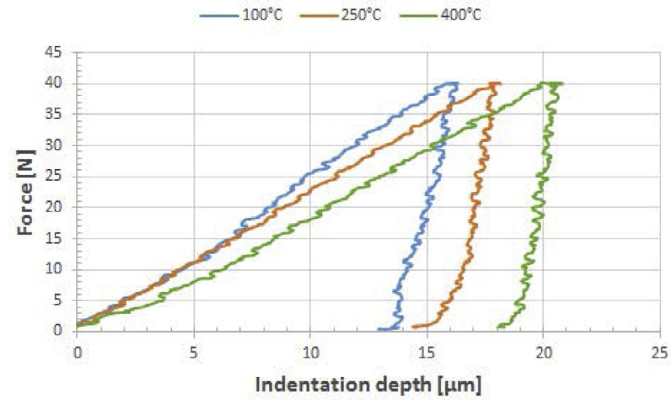

Fig. 4. EUROFER load-displacement-curves for different temperatures, achieved with the spherical diamond tip.

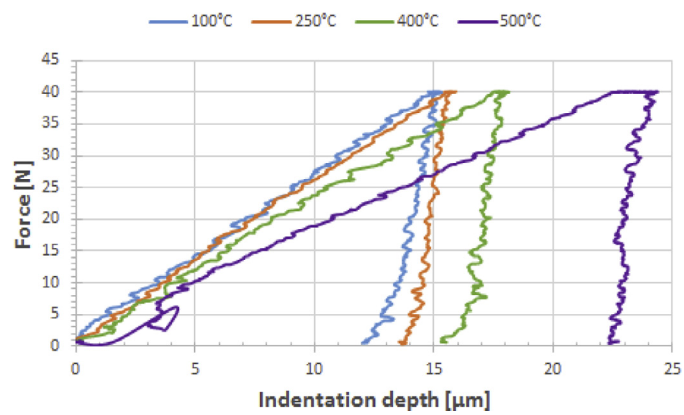

Fig. 5. EUROFER load-displacement-curves for different temperatures with a spherical sapphire tip.

The unsteady behavior at the beginning of the curve at 500 °C is caused by an inaccurate displacement measurement when the first contact between sample and indenter was made. A repeat of the indentation test was not possible, due to the limited size of the sample.

Overall, the results obtained with the diamond tip are more reliable than with sapphire tip. The diamond results illustrate the continuous temperature dependent behavior of the material in a reliable way. In contrast, the sapphire results don't reflect that behavior in the same accuracy, see the curves for 100 °C and 250 °C in Fig. 5. Hence, the result of investigations with diamond as tip material will be used as reference for future evaluations of the experiments with sapphire tips.

Nevertheless, it is not possible to check the quality of the tips after every single indentation, due to the vacuum operation of the device. But in future, after a measurement campaign of a whole sample, a possible tip variation can be detected and correlated to significant discrepancies of the indentation results.

4. Numerical simulation

For a more intense investigation of the influences on the indentation behavior with respect to the tip material and deviations of the tip geometry, simulations are carried out with the software *ABAQUS*.

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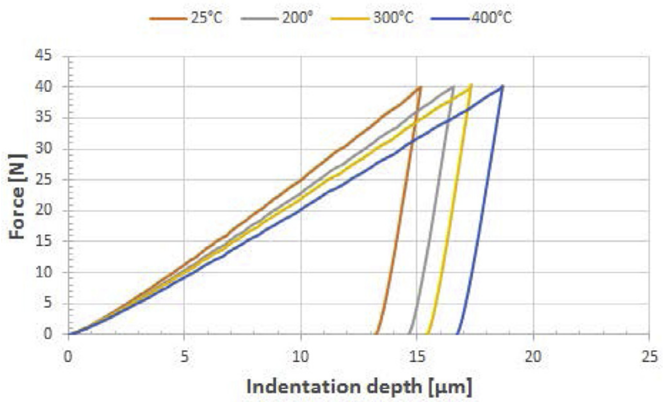

Fig. 6. Calculated load-displacement-curves for different temperatures with a spherical diamond tip (Radius of 200 µm).

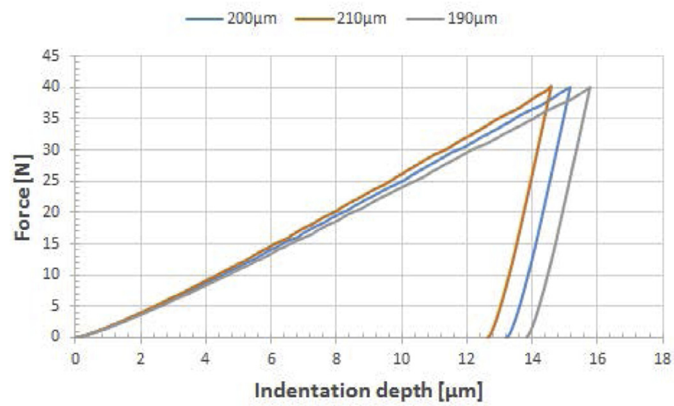

Fig. 8. Calculated load-displacement-curves for different tip radii for a diamond tip at room temperature.

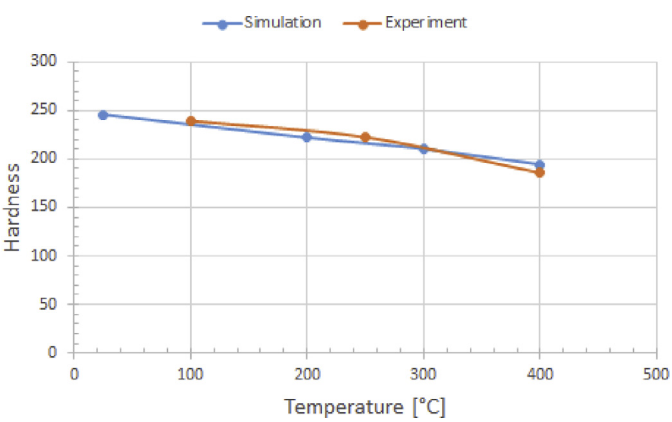

Fig. 7. Brinell hardness determined for the calculated and experimental load-displacement-curves for a spherical diamond tip.

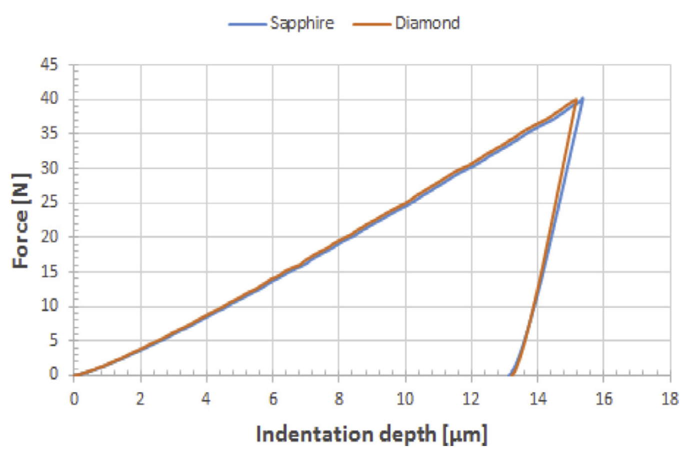

Fig. 9. Calculated load-displacement-curves for different indenter tip materials at room temperature.

For the numerical calculations, the EUROFER material data of the *RCC-MRx* [18] are used. The model consists of two parts, the indenter and the sample. For a reduction of the calculating time, symmetrical boundary conditions are used. For the sample, an elasto-plastic material behavior and for the indenter an elastic material behavior is assumed. No time-dependent behavior was taken into account. Obviously, the indentation depths calculated in this study reflect not only the penetration of the indenter in the sample, but also include the elastic deformation of the indenter tip, as discussed above.

At first, calculations for different temperatures with a diamond tip are carried out. These results show a similar material behavior as the experimental results, a decreasing deformation resistance and an increasing indentation depth with an increasing temperature, see Figs. 4 and 6. For the calculations, the material data of the *RCC-MRx* code were chosen from the relevant temperature regime.

In Fig. 7, the hardness vs. the temperature is shown for the calculated load-displacement-curves of Fig. 6 and the experimental curves of Fig. 4. For the hardness comparison of the experimental and simulated data, the indentation depths after load removal are determined and the indentation diameters are calculated by using the formula of a spherical shape. The hardness is calculated according to the Brinell formula.

The hardness values of the simulations show a similar temperature dependency of the tested material as the hardness of the experimental results. The experimental values are slightly higher at 100 °C and 250 °C, and lower at 500 °C. This is caused by not perfect and unsteady unloading parts of the experimental load-displacement curves, see Fig. 4. Here, the lower parts are more sensitive to misinterpretations.

Calculations of the hardness values, on basis of a fitting of the upper parts of the load-displacement-curves, exhibit more comparable results of the experiments and the simulations.

In Fig. 8, calculated load-displacement-curves for different radii of the spherical indentation tips are plotted. For the calculations, tip radii of 190 µm, 200 µm and 210 µm are used. For a decreasing tip radius, the indentation depth increases for the same applied load. The deviation of 10 µm, 5% of the tip radius, causes a variation of indentation depth after unloading of about 4–5%. In contrast, the corresponding diameters of the residual indents only show a variation of about 0.5%. It is noted that variations of 10 µm in tip radius are 50% of the acceptable radius deviation for the production of spherical tips according to DIN EN ISO 14577-2 and are in the range of the accuracy of the tip production. Hence, the dimensions of the variations which are shown in Fig. 8 have to be kept in mind during the evaluation of the experimental results.

Additionally, the influence of the elastic behavior of the tip material is investigated. In Fig. 9, the calculated load-displacement-curves for sapphire and diamond as tip material are shown. The indentation depth of sapphire is 1.3% higher than for diamond, due to the different Youngś modulus of the materials. Also, the influence of the tip material on the unloading part of the curves is seen as a smaller slope for the sapphire tip. This is in good agreement with the results of indentation experiments on EUROFER in [15].

The fact that the calculated indentation depth at full load for the sapphire tip is higher than for the diamond tip means that the

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Fig. 10. SEM micrograph of a spherical diamond tip with EUROFER deposition after a high temperature test.

hardness of the tested material is underestimated with the sapphire tip for evaluation methods which use the indentation depth under load, e.g. Martens hardness, DIN EN ISO 14577-1. Here, the plastic and elastic deformations are taken into account.

After the unloading, a slightly smaller indentation depth for the sapphire and hence a smaller residual imprint is obvious. This means, that the material of the tip also has an influence on the diameter of the residual indent and hence on the Brinell hardness. Therefore, a conventional hardness determination with a sapphire tip gives slightly higher values compared to one with a diamond tip.

By comparison of the indentation depths of the indenters used in this study, the influence of a geometrical deviation of the tip, with respect to the production accuracy, is higher than the influence of the tip material. Therefore, it is important to conduct reliable tip shape calibration procedures throughout experimental indentation studies, in particular, at elevated temperatures since the tip shape may change due to the interaction with the sample materials. Nevertheless, a consideration of the lower elastic modulus of tip materials others than diamond is also important for a reliable evaluation of the indentation experiments.

5. Hot Cell operation of the device

For an effective operation of the high temperature indentation device in a hot cell in the Fusion Materials Lab some restrictions have to be taken into account. An easy access to the device is needed for a secure handling of different device parts with manipulators, e.g. the optical measurement system, the positioning of the specimen on the sample stage and the exchange and fixing of the thermocouple of the indenter. Another important challenge is the quality management of contaminated indenter tips. A frequent inspection of the tips is necessary for an identification of defects and hence to ensure a reliable evaluation of the experiments, compare Fig. 10.

All new indenter tips are measured via a laser scanning microscope to determine the tip radius and characterize deviations from the ideal shape caused by inaccuracy of the production procedure. During the operation in a Hot Cell, the indenter surface will be inspected frequently via a scanning electron microscope, and indentation tests on materials with known properties, e.g. fused silica, are carried out. Thus, deviations of the quality of the tips can be verified and considered in the experiment evaluation. Hence, reliable results can be ensured.

6. Conclusion and outlook

In this study, diamond and sapphire are used as tip materials for an investigation of the reduced activation steel EUROFER by instrumented indentation at elevated temperatures. The conventional hardness evaluation according to Vickers on EUROFER clearly shows a dependency on the testing temperature. This corresponds to results of the Brinell hardness values and the measured load-displacement-curves of EUROFER. Between the Vickers tips with different tip materials, diamond and sapphire, a continuous difference of HV was detected over the whole test temperature range, a fact that was observed for spherical tips for investigations on MANET II as well, [15].

The numerical simulations of the indentation procedure deliver a benefit for the evaluation of the experimental data sets. The influence of the tip material can be determined numerically and verifies the experimental results and argumentations stated in [15]. Additionally, the effect of deviations of the radius of spherical tips in a range of 10 µm is quantified.

With respect to the temperature dependency, experimental results and simulations exhibit a similar behavior. The Brinell hardness values calculated on basis of load-displacement-curves show a satisfying correlation of simulations and experiments as well.

In summary, it can be stated that sapphire as tip material has an influence on the measured indentation depth under load and after unloading, because of its lower Youngś Modulus compared to diamond. However, for a practical point of view these differences can be taken into account in the data analysis and are rather small compared to possible imperfections of the tip.

The study has validated the functionality of the high temperature indentation device. It is feasible to create a data base, which will support further investigations, like multi-cyclic indentation tests. With such tests a stress-strain-curve of the tested materials can be determined [19]. It was also demonstrated that the numerical simulations will help to analyze experimental results.

Another goal for the application of the high temperature indentation device is to investigate materials with respect to their ductile-to-brittle transition temperature (DBTT). Materials with a body-centered cubic crystal system exhibit brittle and a ductile behavior depending on the temperature and on the strain-rate of the loading [20]. Hence, indentation experiments at different temperatures with focus on the influence of the loading rate can give a hint to the DBTT. The temperature regime up to 650 °C of the indentation device is principally well suited for an investigation of fusion relevant materials including EUROFER after irradiation and even tungsten. Future experiments will focus on this aspect giving a valuable benefit for the characterization of irradiated materials with respect to the neutron induced embrittlement.

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